High-temperature leaching with the ionic liquid [Hbet][Tf_2N] for the recovery of metal values from NdFeB magnets

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In 2014, the European Commission defined rare earths (REEs) as critical raw materials. [1] The criticality does not arise from their scarcity, but from China's monopoly and the need of the European Union (EU) to become independent from the Chinese market. The EU has a large supply of secondary raw materials in, at least, 150'000 highly valuable landfills. Neodymium iron boron (NdFeB) magnets are currently the most relevant source for RE recycling and trendsetter for dysprosium and neodymium applications.[2]

Ionometallurgy is a pool of techniques based on traditional hydrometallurgy using ionic liquids (ILs) as solvent, instead of mineral acids or traditional organic solvents. ILs are solvents made entirely of ions and they are offering a new coordination environment for metal ions. ILs functionalized with an acidic group can be useful for selective dissolution or leaching of metals from complex solid materials.

To recover neodymium, dysprosium and cobalt from end-of-life (EoL) NdFeB magnets and production scrap, the IL betainium bis(trifluoromethylsulfonyl)imide [Hbet][Tf_2N] was used as leaching agent. The leaching process was carried out at 175 °C to increase the kinetics: this is a new application of ionic liquids, which are mostly used at low temperatures.

The solids were first crushed and milled, to reduce the particles size and increase the surface area. In a scond step, they were then roasted to oxidize the metals, to avoid IL decomposition by reduction and to increase the leaching selectivity. The particles size, composition and structure of the obtained powders were determined. After TGA analysis of the [Hbet][Tf_2N] ionic liquid, the leaching temperature was set at 175 °C. The effect of the leaching time and particles size on the leaching of NdFeB magnets and scrap with dried [Hbet][Tf_2N] was investigated.

Low recovery efficiencies (< 10%) and selectivities of the valuable metals were achieved. On the contrary, dissolution of pure metal oxides (Nd_2O_3, Dy_2O_3 or Fe_2O_3) in [Hbet][Tf_2N] at 175 °C resulted in recovery efficiencies close to 100%. The roasted magnets contain $FeNdO_3$, which is more stable than Nd_2O_3 or Fe_2O_3 under the same conditions. The leaching process developed was compared to one of Dupont and Binnemans, in which they leached 100% REEs and cobalt from EoL NdFeB magnets with water-saturated [Hbet][Tf_2N] at 90 °C. [3] The effect of water content on both mass transfer and metals coordination was investigated. The mass transfer is not hampered as the viscosity of dried [Hbet][Tf_2N] at 175 °C was found to be very similar to the viscosity of water-saturated [Hbet][Tf_2N] at 90 °C. The presence of water was proved to be crucial for metals coordination. Dried [Hbet][Tf_2N] is not able to saturate the coordination sphere of the rare-earth ions in contrast to water-containing [Hbet][Tf_2N], thus the metals are more stable in the solid phase.[4]

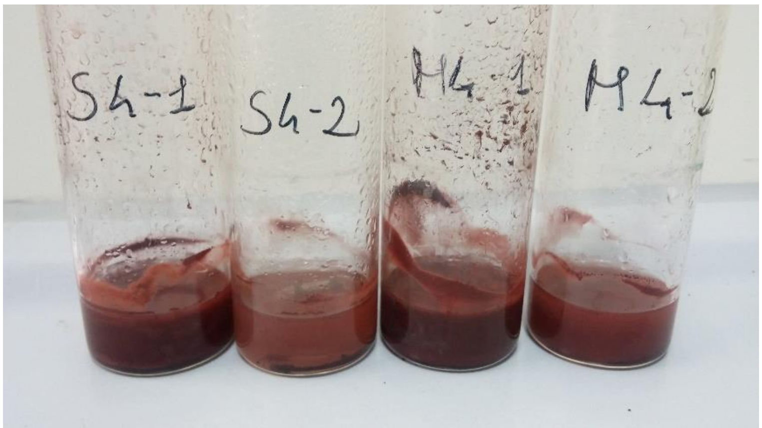

Fig. 1: Leachates from milled and roasted NdFeB magnets after 48 hours of leaching at T = 175 °C with [Hbet][Tf_2N].

[1] Report on Critical raw materials for the EU, European Commission, DG Enterprise & Industry; Brussels; 2014

[2] G. P. Hatch; "Rare-Earth Re-Use and Recycling"; NATO STO AVT-285 Lecture Series: Rare Earths – Securing Supply Chains, Materials and Technologies; 2017

[3] D. Dupont and K. Binnemans; Green Chem. 17; 2150–2163; 2015

[4] P. Nockemann et al. ; *J. Phys. Chem. B.*; 110; 20978-20992; 2006